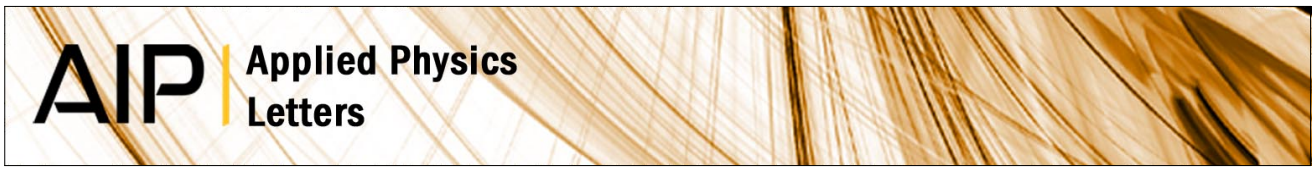

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Spectral phase-matching properties for second harmonic generation in nonlinear crystals

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The spectral phase-matching bandwidths in $LiIO_3$, $LiNbO_3$, CDA, and KDP (type I and type II) for second harmonic generation (SHG) at 1.06 μm have been accurately measured using a tunable line-narrowed Nd:glass laser. The present results disagree with previously published data. Mechanisms such as sum-frequency generation which could contribute to the discrepancy are discussed and demonstrated in a 90° phase-matched deuterated CDA crystal.

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It is known that efficient SHG in nonlinear crystals requires phase matching of the fundamental and the second harmonic wave vectors. The spectral bandwidth for phase matching is restricted by the dispersion properties of the nonlinear crystals.

Using a tunable line-narrowed Nd:glass laser, values of spectral phase-matching bandwidth for second harmonic generation (SHG) in $LiIO_3$, $LiNbO_3$, CDA, and KDP (type I and type II) have been measured at 1.0640 μm. The results disagree with previously published data including that in a review article published in 1975.[1] This disagreement led to consideration of possible mechanisms which may have contributed to the discrepancy.

In this work, the spectral phase-matching bandwidth, $L\Delta\lambda$, of the nonlinear crystals at 1.0640 μm was measured using a tunable Nd:glass laser scanned across the gain bandwidth of the laser, where L is the length of the crystal and $\Delta\lambda$ is the full spectral width of the fundamental at which the second harmonic conversion efficiency for constant fundamental power density reduces to the half-maximum (FWHM). A Pockels-cell Q-switched Nd:glass laser (ED-2 laser glass) was line narrowed with a piezoelectrically driven Fabry-Perot etalon (Burleigh Model TL-15) with 100 μm spacing and 50% reflectivity at 1.06 μm. The laser was operated at 10 mJ, TEM_{00} with 100 nsec duration. The linewidth was 1 A and tunable across a bandwidth of about 100 A. The spectra were analyzed with a Jarrel-Ash spectrometer and imaged with a silicon-vidicon camera (GE Model 4TE26) with an overall resolution of about 0.3 A. The nonlinear crystals were placed in the near field of the resonator without focusing and oriented in such a direction that the maximum SH was generated at the centered wavelength of 1.0640 μm. The SH intensity was measured as a function of fundamental wavelength scanned across 1.0640 μm at a constant fundamental power level. The following crystals were studied: (a) a temperature-tuned 90° phase-matched $LiNbO_3$, (b) a temperature-tuned 90° phase-matched deuterated CDA, (c) an angle-tuned $LiIO_3$, (d) an angle-tuned (type I) KDP, and (e) an angle-tuned (type II) deuterated KDP. These results are shown in Fig. 1 and compared with the theoretical curves, $\mathrm{sinc}x=(\sin x/x)^2$; the agreement is excellent. The result for case (e) is similar but not shown here. The results are summarized in Table I.

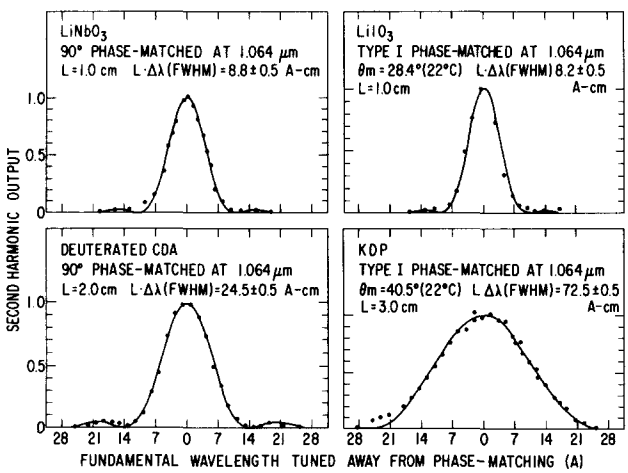

FIG. 1. The spectral bandwidth for second harmonic generation for nonlinear crystals $LiNbO_3$, $LiIO_3$, CDA, and KDP near 1.0640 μm measured with a tunable line-narrowed Nd:glass laser.

The spectral phase-matching bandwidth $\Delta\lambda$ is given by

$$\Delta\lambda(\mathrm{FWHM})=\frac{\lambda_1}{2L}\left(\frac{\partial\eta_1}{\partial\lambda_1}-\frac{1}{2}\frac{\partial\eta_2}{\partial\lambda_2}\right)^{-1}, \tag{1}$$

where η_1, η_2, λ_1, and λ_2 denote the refractive indices and wavelengths at the fundamental and the second harmonic, respectively. Previously, the $L\Delta\lambda$ values for $LiNbO_3$ and KDP were calculated from known dispersion data to be 3.4 and 400 A cm, respectively, at 1.06 μm.[1] The present results disagree with the earlier results. It is believed that the discrepancy is due to the sensitivity of Eq. (1) to the dispersion property of nonlinear crys-

TABLE I. Spectral bandwidth for second harmonic generation in nonlinear crystals at 1.0640 μm.

Crystal	Type of phase match	$L\Delta\lambda$ (FWHM) (A cm)
$LiNbO_3$	90° phase matched	8.8±0.5
CDA (deuterated)	90° phase matched	24.5±0.5
$LiIO_3$	Phase matched at $\theta_m=28.4°$	8.2±0.5
KDP	Phase matched at $\theta_m=41.5°$ (type I)	72.5±0.5
KDP (deuterated)	Phase-matched at $\theta_m=53.5°$ (type II)	55.7±0.5

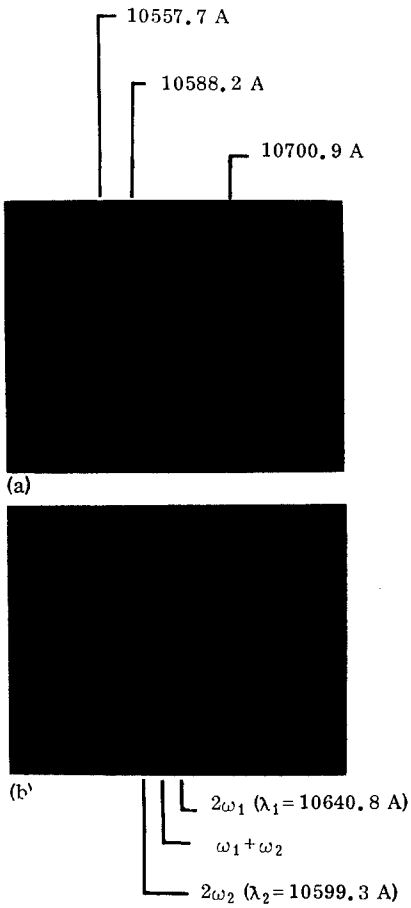

FIG. 2. Second harmonic signals when the fundamental consists of (a) a single frequency and (b) two distinctive frequencies. In (b), both frequencies are outside the half-width for phase matching, and the temperature of the deuterated CDA crystal (1.5 cm long) was set for phase match at $(\omega_1+\omega_2)/2$. The thallium 3529 Å and 3519 Å doublet lines in third order and 5350.5 A line in second order were used for calibration and are shown in the upper record of each pair.

tals which makes difficult the accurate calculation of the spectral phase-matching bandwidth.

Furthermore, the present results also disagree with several earlier observations[1] using a broadband Nd:glass laser as the fundamental source. It is shown below that the phase-matching constants such as $L\Delta\lambda$ can be grossly overestimated by use of a relatively broadband source such as the unnarrowed Nd:glass laser. Such a broadband fundamental source introduces an additional mechanism for production of the second harmonic, namely, sum-frequency generation through parametric interaction between symmetrically displaced spectral components. The interaction between such symmetrically located spectral components, each of which itself can lie far outside the phase-matching bandwidth, can be strong. As a result, the apparent spectral phase-matching bandwidth for SHG can be increased. A similar situation, the determination of nonlinear susceptibility in the presence of multimode structure in a laser, has been noted by Bloembergen.[2]

Consider a nonlinear crystal at a given orientation such that the fundamental wave vector $\mathbf{k}_0$ is phase matched for SHG, namely, $\mathbf{k}_1 = 2\mathbf{k}_0$ and $\omega_1 = 2\omega_0$. In a broadband source, those wavevector pairs $(\mathbf{k}_2, \mathbf{k}_3)$ satisfying $|\mathbf{k}_2 - \mathbf{k}_0| \gg \frac{1}{2}\Delta k$ and $|\mathbf{k}_3 - \mathbf{k}_0| \gg \frac{1}{2}\Delta k$ can interact strongly as long as $|\mathbf{k}_2 + \mathbf{k}_3 - 2\mathbf{k}_0| < \Delta k$, where Δk is the full width for wave-vector phase match for SHG at $\mathbf{k}_0$ at the given crystal orientation. This kind of parametric sum-frequency generation was unambiguously demonstrated in a 90° phase-matched deuterated CDA crystal, described next.

For this measurement, two line-narrowed fundamental frequencies at ω_1 and ω_2 of equal intensity were generated simultaneously from a Nd:glass laser and brought into a temperature-tuned 90° phase-matched deuterated CDA crystal. The spectral spacing between these two fundamental frequencies was determined by the free spectral range of a $LiNbO_3$ birefringent intracavity tuning element[3] and was chosen to be sufficiently greater than the phase-match bandwidth given in Table I so that no two frequencies among ω_1, $\frac{1}{2}(\omega_1+\omega_2)$, and ω_2 could be simultaneously phase matched at any given temperature.

In Fig. 2, the upper record in each pair shows the calibrating wavelengths, and in the lower records the second harmonic signals are shown spectrographically when the incident beam consists of (a) a single frequency and (b) two frequencies at ω_1 and ω_2. The crystal was temperature tuned to phase match at the single laser frequency for case (a), but midway between the two laser frequencies for case (b), i.e., at the center frequency $\frac{1}{2}(\omega_1+\omega_2)$. In the latter case, the second harmonic signal at $(\omega_1+\omega_2)$ was overexposed to let much weaker signals at $2\omega_1$ and $2\omega_2$ be visible.

In the experiment which produced the results of the top trace in Fig. 3, two laser lines were again generated at ω_1 and ω_2, and the second harmonic signal was recorded as the temperature of the crystal was continuously scanned, causing the crystal to phase match at ω_1, $\frac{1}{2}(\omega_1+\omega_2)$, and ω_2 sequentially at three different temperatures and producing the three peaks shown.

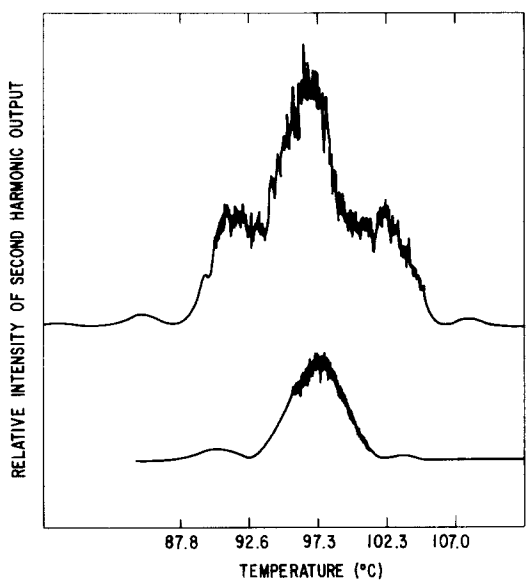

FIG. 3. The temperature signatures for the cases of a single line (lower) and a pair of lines (upper) as in the case of Fig. 2.

This is to be compared with the bottom trace in which the second harmonic signal is shown when the crystal temperature is scanned through phase match with an incident beam that has only one frequency component.

The top trace consists of three superimposed sinc$x$ functions corresponding to the second harmonic signal at $2\omega_1$ and $2\omega_2$ and the sum frequency at $(\omega_1 + \omega_2)$. In this case, the two incident frequencies were of about equal intensity, and the separation was about 42 A, which is large compared with our measured value $\Delta\lambda$ for a 90° phase-matched deuterated CDA crystals 1.5 cm long. Note that the relative intensity in the top trace of Fig. 3 shows that the sum-frequency intensity is proportional to the integrated square of the fundamental intensity not to the square of integrated fundamental intensity. From the top trace of Fig. 3, one can also obtain the following constants for the 90° phase-matched deuterated CDA crystal: $L\Delta T(\text{FWHM}) = 6.06$ °C cm $d\lambda_m/dT = 0.3$ A/°C, where ΔT (FWHM) is the temperature bandwidth for phase match and λ_m is the center wavelength for phase match. Furthermore, from the measured values of $\Delta\lambda$, $\Delta\theta$, and ΔT, one can obtain a series of derivatives such as $\partial\theta/\partial\lambda$, $\partial T/\partial\lambda$, etc.

In summary, the spectral phase-matching bandwidths in several of the most useful nonlinear crystals at 1.06 μm have been measured accurately using a tunable line-narrowed Nd:glass laser. The discrepancies between this work and previously published data are discussed and mechanisms which could produce the discrepancies are suggested. The observation of sum-frequency generation in a 90° phase-matched deuterated CDA crystal demonstrates the inadequacy of using a broadband source to determine nonlinear phase-matching constants.

The author wishes to thank Dr. Jack Wilson of Laboratory for Laser Energetics at the University of Rochester for loan of the KDP type I crystal.

[1]Previously, from the published literature, the spectral phase-matching bandwidth for SHG was determined by several approaches: (a) those calculated by known dispersion properties of the nonlinear crystals, such as R.C. Miller [Phys. Lett. A 26, 177 (1968)] and S.A. Akhmanov, A.I. Kovrygin, and A.P. Sukhorukov [in *Quantum Electronics, a Treatise*, edited by H. Rabin and C.L. Tang (Academic, New York, 1975)]; (b) those determined with a broadband source such as J. Comly and E. Garmire, Appl. Phys. Lett. 12, 7 (1968); S.L. Shapiro, Appl. Phys. Lett. 13, 19 (1968); A.J. DeMaria, W.H. Glenn, M.J. Bvierza, and M.E. Mack, Proc. IEEE 57, 1 (1967); W.H. Glenn, IEEE J. Quantum Electron. QE-5, 284 (1969). There was a measurement on CDA using a tunable Nd:glass laser reported by D.J. Taylor [J. Appl. Phys. 46, 3988 (1975)] and his result agrees with present one. Another measurement using a two-frequency line-narrowed Nd:glass laser was reported by R.B. Andreev, V.D. Volosov, and A.G. Kalintsev [Opt. Spectrosc. 37, 169 (1974)]. Their results differ considerably from present ones.

[2]N. Bloembergen, *Non-linear Optics* (Benjamin, New York, 1965).

[3]Y.S. Liu, J. Appl. Phys. 48, 647 (1977).

Transient versus steady-state molecular absorption: Application to CO_2 laser pulse-duration discrimination[a)]

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Transmission of short CO_2 laser pulses in heated CO_2, including pulse reshaping and ringing, is studied using a single-sweep oscilloscope-detector combination having a 100-psec rise time. We demonstrate that hot CO_2 is extremely effective in increasing by as much as 1000-fold the pulse-to-background intensity ratio in short-pulse CO_2 laser systems.

PACS numbers: 42.10.−s, 42.80.Cj, 42.60.He, 42.55.Dk

We report experimental and theoretical studies on nonexponential attenuation of short pulses in narrow-band absorbers. These studies lead directly to a novel method for improving the contrast ratio (pulse-to-background intensity) of a subnanosecond CO_2 laser pulse externally gated from a long temporally smooth pulse.[1] This improvement is necessary in large laser-fusion systems because as little as 50 μJ of amplified feedthrough (oscillator radiation prior to the desired pulse) can damage fusion targets.[2] Feedthrough is especially troublesome in multifrequency subnanosecond CO_2 systems characterized by large linear gain and saturated operation.

In some systems, the short pulse to be injected into the amplifier chain is electro-optically switched out[3] of a longer temporally smooth pulse. A closed electro-optic switch unfortunately leaks about 10^{-3} of the 10.6-μm beam. This long pulse feedthrough, weak and narrow band, experiences the amplifier chain's peak unsaturated gain and may grow to contain an appreciable fraction of the output energy. Previous attempts to minimize 10.6-μm oscillator feedthrough have included multiple electro-optic shutters[4] using Pockels effects, and solid-state[5] and gaseous[6] saturable absorbers.

a)Work performed under the auspices of the U.S. ERDA.